7th Int. Symp. "Nanostructures: Physics and Technology"
St Petersburg, Russia, June 14–18, 1999

Optimization of the size distribution of self-organized quantum dots

S. Bose and *E. Schöll*
Institut für Theoretische Physik, Technische Universität Berlin,
Hardenbergstr. 36, 10623 Berlin, Germany

Abstract. We have performed kinetic Monte Carlo simulations which explain the self-organized Stranski-Krastanov growth of quantum dots by the nonlinear interplay of deposition, surface diffusion, and the strain field. The optimization of the size distribution and the spatial arrangement of the quantum dots is discussed with respect to the coverage of the semiconductor surface.

The self-organized growth of quantum dots in strained semiconductor systems has recently become the focus of extensive research because of its great potential for application to nanoelectronic devices. It might help to simplify the fabrication of novel semiconductor devices based on quantum dots, for instance quantum dot lasers. It is widely accepted that the key for understanding this growth lies in the strain fields which are present in the Stranski-Krastanov growth mode [1].

Here we consider the initial stage of growth of quantum dots in regular arrays with a sharp size distribution. While the detailed shape of the fully developed quantum dots [2] is mainly governed by the specific materials the quantum dots are made of, the self-ordering in regular arrays, with sharp size distribution, appears to be a universal feature which is found in the technologically most relevant III-V semiconductor compounds, like InAs/GaAs(001) [3], but has also been observed in the Ge/Si(001) system [4, 5, 6] as well as in Ag/Pt(111) [7]. A theoretical approach based on thermodynamic equilibrium considerations was developed by Shchukin et al. [8].

In this contribution we use a dynamic approach based on kinetic Monte Carlo simulations to discuss the optimization of growth conditions. The main idea is to introduce a strain field around the quantum dots which should have two effects. First, it should destabilize the boundary of the island at which it is centered, similar to the mean field theory by Dobbs et al. [9] or other Monte Carlo simulations by Ratsch et al. [10] or Barabasi [11]. Second, the long range effects of the strain field should provide a medium for the coordination of the quantum dot configuration on the surface. Besides the standard energy terms for the surface binding energy of an atom E_S and the binding energy to a nearest neighbor E_N we introduce an energy correction term due to the strain field $E_C(x, y)$ which depends on the coordinates x and y on the surface, because the strain is high near large islands and decays quite fast with increasing distance from the islands. This correction term depends linearly upon the underlying strain field [12]. The probability for a single atom to jump from one lattice position to another is now given by

$$p = f \exp\left[-\frac{E_S + nE_N + E_C(x, y)}{k_B T}\right] \tag{1}$$

where f is the frequency of attempts per second, n is the number of nearest neighbors, k_B is Boltzmann's constant and T is the temperature.

Due to the strain the atoms are shifted from their equilibrium positions which generally leads to a reduction in binding energy. Therefore $E_C(x, y)$ is a negative quantity. To model

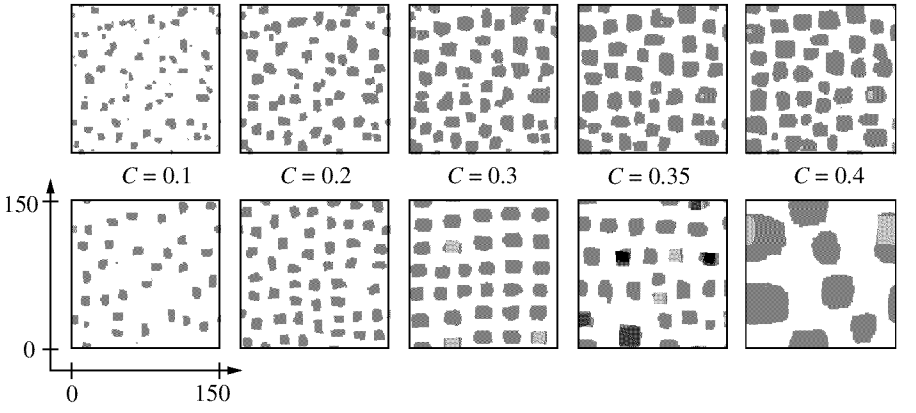

Fig. 1. Monte Carlo simulations of the structure of the surface for different coverage C right at the end of the deposition (upper row) and after 50 s (lower row). The simulation was performed with a growth rate of 0.1 ML/s at 750 K. Atoms in the first and the second monolayer are indicated by dark and light shading, respectively.

the spatial dependence of $E_C(x, y)$ around a single island, we use a phenomenological function which decreases linearly from the boundary of the island, is centered at its center of mass, and has a cubic symmetry and an amplitude depending upon the size of the island. The following parameters are used for the simulations: $E_S = 1.3$ eV, $E_N = 0.3$ eV [ ]. The amplitude of the strain field grows by a factor of 0.001 eV per atom and has a range of three times the effective "radius" of the island. We also incorporate a Schwöbel barrier $E_{\mathrm{Sch}} = 0.1$ for atoms jumping up to the next monolayer.

We have performed Monte Carlo simulations with a constant flux of atoms for different temperatures, deposition rates, and growth interruptions. If the temperature is fixed to 750 K and the growth rate is chosen as 0.1 monolayers(ML)/s we get the following sequence of images shown in the upper row of Fig. 1 if we interrupt the deposition at different coverage C. With a growth rate of 0.1 ML/s we reach a coverage of 10%, i.e. 2250 atoms on a 150×150 lattice, after 1 s, a coverage of 20% after 2 s and so on. From the structure of the surface we can extract the size distribution of the islands (quantum dots) which is shown in the upper row of Fig. 2. In the histograms the number of islands is plotted versus the square root of the number of atoms in a dot which is a measure of the mean base length or lateral diameter N of a square island.

It is obvious that at the end of the deposition time the array of self-assembled quantum dots is not of high quality. The size distribution is always broad and the spatial arrangement of the quantum dots on the surface is quite irregular. But there is another notable result. The maximum of the size distribution increases with increasing coverage from about $N = 5$ at $C = 0.1$ to about 15 at $C = 0.35$ beyond which it does not increase any further. This value marks the point where the destabilizing effect of the strain prevents further lateral growth of a single island and the transition from 2D to 3D islands can take place.

During this first stage of the growth a constant flux of atoms keeps the system far from thermodynamic equilibrium. After the growth is interrupted, the simulation is continued without flux; thus the system will evolve towards some state of relative, not necessarily absolute minimum energy. The simulation is stopped after a total simulation time of 50 s. The resulting structures and the corresponding size distributions are shown in the lower

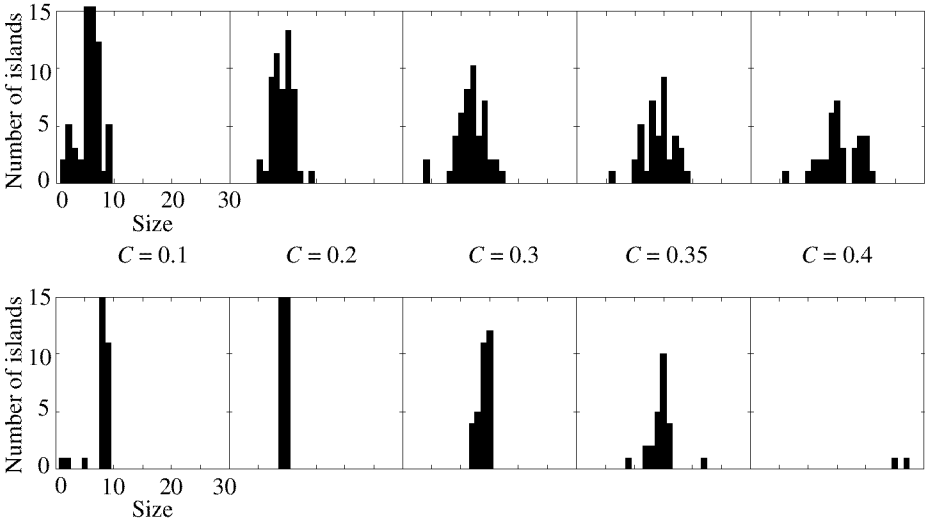

Fig. 2. The distribution of islands sizes for the surface structures shown in Fig. 1.

rows of Fig. 1 and Fig. 2, respectively. Now the different kinds of ordering are clearly visible. The size distribution always becomes sharply peaked, except for $C = 0.4$, and the maximum moves to larger sizes with increasing coverage up to a maximum size of about $N = 15$. The spatial ordering is best at $C = 0.3$, but there are regions of local ordering for $C = 0.2$ and $C = 0.35$ as well. For $C = 0.1$ and $C = 0.2$ the islands are not large enough to interact, and at $C = 0.35$ there occurs already some clustering of islands which spoils the spatial pattern. For a coverage of $C = 0.4$ the islands are completely clustered. The transition from 2D to 3D growth starts at $C = 0.3$ where some islands already consist of two monolayers.

These results show that it is easier to obtain a sharp size distribution than a perfect spatial ordering. There are two reasons for that. First, the interaction which leads to the uniformity of the island sizes is a "long-range" interaction. For example, in the system with a coverage of $C = 0.1$ we have no spatial ordering but a sharp size distribution. The islands are too far apart and too small to interact directly to form a regular spatial pattern, but the destabilizing effect of the strain is stronger for larger islands than for smaller ones, and hence atoms are more likely to move from a large island to a smaller one than vice versa. This consequently leads to approximately equal island sizes. Second, the formation of patterns is a more local effect. If the critical island size and the corresponding critical density for the island-island interaction is reached, the islands start to align locally. If two neighboring islands are too close they will tend to move apart slowly, but sometimes this may be impeded by other close-by islands. It sensitively depends upon how and where the islands nucleate if it is possible for an island to move to a position where it will have the same optimum distance to all its neighbors. In small local regions, the pattern of quantum dots might be very regular, but in larger regions more and more defects may occur in the pattern.

In conclusion, we have presented results of a kinetic Monte Carlo simulation of the initial growth stage of quantum dots. Two different kinds of self-ordering, namely the ordering of the sizes of the dots and their lateral arrangement on the surface, were discussed in

dependence on different surface coverage. Our simulations predict a sharp size distribution up to a critical coverage where the islands start to form clusters. Spatial ordering sets in shortly before the clustering occurs. It has always been found necessary to let the system evolve freely for some time after the end of the deposition to achieve strong ordering.

References

[1] D. Bimberg, M. Grundmann and N. Ledentsov, *Quantum Dot Heterostructures* (John Wiley & Sons Ltd., New York), 1999.

[2] N. Moll, A. Kley, E. Pehlke and M. Scheffler, *Phys. Rev. B* **54**, 8844 (1996).

[3] N. N. Ledentsov, M. Grundmann, et al., *Solid State Electron.* **40**, 785 (1996).

[4] D. J. Eaglesham and M. Cerullo, *Phys. Rev. Lett.* **64**, 1943 (1990).

[5] G. Abstreiter, P. Schittenhelm, C. Engel, E. Silveria, A. Zrenner, D. Meertens and W. Jäger, *Semicond. Sci. Technol.* **11**, 1521 (1996).

[6] O. G. Schmidt, C. Lange, K. Eberl, O. Kienzle and F. Ernst, *Appl. Phys. Lett.* **71**, 2340 (1997).

[7] R. Schuster, H. Röder, K. Bromann, H. Brune and K. Kern, *Phys. Rev. B* **54**, 13476 (1996).

[8] V. A. Shchukin, N. N. Ledentsov, M. Grundmann and D. Bimberg, in *Optical Spectroscopy of Low Dimensional Semiconductors*, *NATO ASI*, edited by G. Abstreiter (Kluwer, Amsterdam, 1997).

[9] H. T. Dobbs, D. D. Vedensky, A. Zangwill, J. Johansson, N. Carlsson and W. Seifert, *Phys. Rev. Lett.* **79**, 897 (1997).

[10] C. Ratsch, P. Šmilauer, A. Zangwill and D. Vvedensky, *J. Phys. I* **6**, 575 (1996).

[11] A.-L. Barabási, *Appl. Phys. Lett.* **70**, 2565 (1997).

[12] E. Schöll and S. Bose, *Solid State Electron.* **42**, 1587 (1998).

[13] S. Clarke, M. R. Wilby and D. D. Vvedensky, *Surf. Sci.* **255**, 91 (1991).